THESIS

EFFECTS OF FUEL MOISTURE CONTENT ON POLLUTANT EMISSIONS FROM A ROCKET-ELBOW COOKSTOVE

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ABSTRACT

EFFECTS OF FUEL MOISTURE CONTENT ON POLLUTANT EMISSIONS FROM A ROCKET-ELBOW COOKSTOVE

Cookstoves have been studied in recent decades for their health- and environment-relevant emissions. Many pollutants, stoves, and burning parameters have been investigated across these studies, including fuel moisture content, which is believed to have substantial impact on stove emissions. Yet, the effects of fuel moisture content on emissions remain poorly characterized.

To address this gap in knowledge, this study characterized particle and gas-phase pollutant emissions during a laboratory experiment exploring three levels of fuel moisture from a single tree sample. Moisture levels tested here varied from 5% to 30% water content, by weight. A novel technique for re-moisturizing the fuel samples was developed and employed to expedite the experimental duration and to ensure consistency across tests.

Results from the study demonstrate strong trends in emissions related to changes in moisture content. Results also suggest there are benefits to drying wood to below 10% moisture content as compared to burning wood that is slightly above the 20% level recommended by the U.S. Environmental Protection Agency. When wood was dried to 5% instead of 25%, modified combustion efficiency improved and average mass-based emissions factors decreased for all pollutant species (formaldehyde, acetaldehyde, benzene, toluene, ethylbenzene, m+p-xylenes, o-xylenes, PM_{2.5}, methane, carbon monoxide, and organic carbon) save black carbon. Dry fuel generated less smoke and higher temperatures than wet fuel. Wet fuel was also difficult to keep lit and burned much slower than dry fuel. Efficiency, burn rate, and stove temperature all affect the way people use their stoves for heating or cooking. Since moisture content impacted efficiency, burn rate, and temperature during this study, as well as multiple gas and particle-phase pollutants, moisture content should be accounted for in future stoves studies, both in the lab and in the field.

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LIST OF NOMENCLATURE

BTEX – Volatile organic compounds: benzene, toluene, ethylbenzene, m+p-xylenes, and o-xylenes

 CH_4 – methane

CO - carbon monoxide

CO₂ – carbon dioxide

EC – Elemental Carbon

EF – Emissions Factor

MC – Moisture Content

MCE – Modified Combustion Efficiency

NDIR - nondispersive infrared (type of sensor technology for detecting gases)

 NO_x – oxides of nitrogen

OC - Organic Carbon

PAH – Polycyclic Aromatic Hydrocarbon

 $PM_{2.5}$ – Particulate Matter under 2.5 micrometers in diameter

PM₁₀ – Particulate Matter under 10 micrometers in diameter

ppmv – parts per million by volume

VOC – Volatile Organic Compounds

CHAPTER 1. INTRODUCTION

1.1 Cookstove Emissions

The 2016 Global Burden of Disease study estimated that household air pollution caused over 2.5 million annual premature deaths globally and was the eighth leading cause of premature deaths behind preventable factors such as smoking, high BMI, and alcohol use [1]. Most household air pollution is produced by primitive stove technologies that are used for cooking and heating homes in low- and middle-income countries. Pollutant emissions from these stoves can also have negative environmental impacts by contributing to greenhouse gases and carbonaceous aerosols in the atmosphere [2, 3].

Many studies have been conducted to understand the underlying mechanisms controlling pollutant emissions from cookstoves; however, few studies have examined fuel moisture content as a key determinant of emissions [2-11]. Studies that have considered moisture have shown varied results in terms of the effects of moisture content on emissions; the results of several key studies are summarized in Table 1.

For example, Chomanee et al. found that increasing the moisture content of rubber wood from 37% to 73% significantly increased concentrations of polycyclic aromatic hydrocarbons (PAHs) and the amount of smoke within a factory [11]. Shen et al. reported that emissions factors (EFs) for particulate matter (PM), organic carbon (OC), and PAHs all increased when moisture content was increased from 5% to 27% [9]. Kinsey et al. found decreases in total particle mass emissions factors when Douglas fir wood was dried from 22% to 12% [12]. Yuntenwi et al. found decreases in mass-based emissions factors of PM_{2.5} and carbon monoxide (CO) at low power for open fire stoves when going from 5% to 15% and increases when going from 15% to 25% [5]. L'Orange et al. found similar decreasing-then-increasing trends of PM_{2.5} and CO emissions for a wood-burning stove as moisture content was varied [13].

Table 1: Literature Review of Fuel Moisture Content Effects on Pollutant Emissions

Author (Year) [Reference]	Fuel Type	Stove(s) Tested	Moisture Range (%)	Study Type	Factors Correlated to Moisture	Correlation when Moisture 个
			, ,		PM EF (g per kg fuel)	↑
					OC EF (g per kg fuel)	↑
Shen (2013)	Poplar	Duial. Caaliina Ctarra	F 2 24	ما ما ما	EC EF (g per kg fuel)	=
[9]	wood	Brick Cooking Stove	5.3 - 31	Lab	Parent PAHs	↑
	logs				Nitrated PAHs	↑
					Oxygenated PAHs	↑
		Top-lit Up-draft (natural draft) Stove	5.9 - 22.1	Lab	Burning rate	\
					Cooking Power	\
					Time to Boil	↑
Huangfu (2014)	Wood Pellets				Thermal Efficiency	↑ then ↓ past 18% MC
[10]	Pellets				CO EF (mg per MJ delivered)	\
					PM _{2.5} (mg per MJ delivered)	\
		Open Fire, Chinese	5 – 30		High Power CO EF (g per kg fuel)	↑ (open fire and skirt), ↓ then ↑ (rocket)
Yuntenwi	Douglas				Low Power CO EF (g per kg fuel)	Varied by stove
(2008) [5]	Fir Sticks	Rocket, Skirt (Open- Walled VITA)	(wet basis)	Lab	High Power PM EF	↑ (skirt), ↓ then ↑
[5]	Sticks	walled vitA)	Dasisj		(g per kg fuel)	(open fire), ↓ (rocket)
					Low Power PM EF	\downarrow then \uparrow (open fire),
					(g per kg fuel)	↓ (rocket and skirt)
					Time to Boil	<u> </u>
				Lab	PM10 EF	↑
N. 4					(g per kg fuel)	•
Magnone (2016)	Oak	Koem wood stove (model KW-1731)	10.34 - 56.31		Organic Carbon EF (g per kg fuel)	1
[3]					Elemental Carbon EF (g per kg fuel)	↑
		Royal Thai and			Thermal Efficiency	V
Bhattacharya	Pine Tree Wood Chips	Forestry Department Stove, Indian "Harsha" Stove, Vietnamese Traditional Stove	9.5 - 24.5	Lab	CO EF (g per kg fuel)	^
(2002)					CO2 EF (g per kg fuel)	V
[4]					(Nitrogen oxides) NOx EF (g per kg fuel)	\
					CH4 EF (g per kg fuel)	П
					MCE	^
		Envirofit G3300		Lab	Fuel Burn Rate	→
	Pod				CO EF (g per kg fuel)	→
Jetter (2012)	Red Oak Sticks		9.2 - 26.2		PM _{2.5} EF (g per kg fuel)	→
[2]*					CO2 EF (g per kg fuel)	↑
					CH4 EF (g per kg fuel)	\
					ultrafine particles (#	↑
					per kg fuel)	<u> </u>
Chomanee	Rubber	ribbed smoked	37.4 -		Total PAHs	↑
(2009) [11]	Wood	sheet, community- level factory oven	73.6	Field	Particle Concentration wed for this study since the	↑

^{*} Jetter et al. tested many stoves and fuels. Only the Envirofit was reviewed for this study since the Envirofit was used in this study and this allowed for better comparison of results to published literature.

With some studies in Table 1 showing significant differences in emissions between moisture levels and others finding insignificant effects or different trends altogether, there is a need to improve our understanding of the magnitude and direction of effects of fuel moisture content on emissions. Equilibrium moisture content (EMC) is the stable moisture content of wood at a set relative humidity and temperature. EMC varies by season and geographic location, such that air-dried wood will reach a range of different moisture contents across the planet [14]. EMCs for various wood species range from below 4 percent in very warm, dry regions to over 20 percent in cool, humid locations. Thus, if other aspects of stove combustion were identical (stove, feed rate, wood species, time of year, etc.), burning the same fuel in Madras, India (10% EMC) and in Lagos, Nigeria (17.6% EMC) could produce different emissions profiles. The EPA recommends burning wood at below 20 percent moisture content but provides no easily accessible explanation as to why this should be done, besides stating that wood burns most efficiently at moisture levels of 15 to 20 percent [15]. Testing low-end moisture levels allowed exploration of the question, "are pollutant emissions reduced by drying fuel completely (say to 5 percent,) as opposed to drying to the EPA recommended 20 percent level?".

1.2 Objectives

This project aimed to isolate and examine the effects of fuel moisture content on the emissions from a rocket-elbow cookstove. Compared to moisture levels tested in previous studies (Table 1), tests were conducted at the lower end of the moisture spectrum. This study examined a wide range of emissions — including volatile organic compounds, carbonyls, gases, and particle matter size, mass, and composition. Analyzing such a range allowed for comparison of results to multiple other studies, listed in Table 1, and allowed for a more comprehensive view into the effects of fuel moisture content on cookstove emissions.

CHAPTER 2. METHODS

1.3 Experimental Design

To gain an understanding of the effects of fuel moisture content on cookstove emissions, three target moisture levels were tested: 5% (low), 15% (medium), and 25% (high) by mass. These levels were selected to examine if there was any benefit to drying wood to below the EPA recommendation of 20% moisture. Two fuel shapes were tested to observe if any differences arose from splitting versus milling the wood. Split wood introduced higher variability into the test procedure, which might more closely resemble variability in tests performed in field studies [16]. Consistency between the shapes of each piece of milled wood, on the other hand, more closely reflects the lower variability of stove tests performed in the lab. Each moisture level/fuel shape combination was tested 4 times, in random order. Four tests were conducted on each of the eight test days. One of the tests every day was a background test (Table 2).

Table 2. Experimental Variables

Stove	Fuel Type	Fuel Shape	Moisture Content	# of Useable Replicates
	Colorado Douglas Fir	Milled (to represent "lab-type" fuels)	5% (Low)	4
			15% (Medium)	4
Envirofit G3300			25% (High)	4
Rocket-Elbow		Split (to represent "field -type" fuels)	5% (Low)	4
			15% (Medium)	4
			25% (High)	3
Background				8

Background tests were conducted to estimate pollutant concentrations that may be present in laboratory background air, so that measured emissions data could be corrected for potential interferences. Background tests were randomly performed during one of the four daily test time slots to avoid systematic errors in correcting for ambient concentrations. Background tests involved setting up all instruments and experimental variables in the exact same manner as the wood-burning tests (except that

no wood was burned). Background tests were run for approximately 1 hour to mimic a normal test duration. No stove was set up and no burning was conducted during these times. Any uncontrollable events, such as tests being performed in the adjacent hood, were carefully noted and explored during data analysis to understand and potentially correct for interference with the emissions levels collected during testing.

The test matrix was randomized to eliminate systematic testing errors. Table 3 in the Appendix displays the randomized matrix. To isolate moisture content in these experiments, all other factors were controlled to the greatest level possible. Only one stove, an Envirofit G-3300 Rocket Elbow Stove, was used for burning fuel. The stove was cooled completely between tests performed on the same day. All tests were started in the same manner and collection of emissions ended before the last test pieces burned out. To remove the influence of startup and shutdown phases on measured emissions, air sampling equipment was turned on after the first set of wood pieces were added, ignited, and shims were almost burned out and equipment was turned off before the last set of fuel pieces were burned out.

1.4 Fuel Preparation

All fuel samples were taken from the same section of a single Colorado Douglas fir tree, with a random selection of heartwood and sapwood included during testing. The tree was sourced from the Colorado State University Forestry Department and was felled during the spring of 2016, approximately 1 year prior to the start of the testing campaign. Half of the tree section (lengthwise) was sent out to be milled with no finish and the other was split with a wood splitter, as depicted in Figure 1.

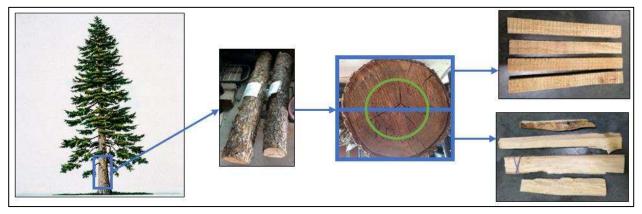


Figure 1. Process of obtaining split and milled fuel pieces from a single Douglas fir tree. Wood inside the green circle is heartwood and wood outside the green circle is sapwood. Both heartwood and sapwood were retained for testing.

By the time testing began, all wood pieces had reached equilibrium moisture content, which is between 6-8% in Colorado [17]. This is well below the fiber saturation point of Douglas fir, the moisture content at which all free water has left the wood but cells are still filled with bound water, which sits at about 26% [18]. Below the fiber saturation point, the wood swells and shrinks as it gains or loses water [18].

Once processing was completed, pieces were placed into bins and six pieces of each type per test were randomly selected before each test day to be used as test fuel while an additional piece of milled fuel was used as the reference piece (the piece to be kiln dried to calculate original moisture content). The day before testing, original weights of each individual piece of wood were measured to 0.1 g precision on a lab scale [NewClassic MF, Mettler Toledo] and recorded. One piece of split wood was then placed into a kiln [National Appliance, 550 W, Model #5830-4] at approximately 105 °C to dry overnight. The rest of the selected pieces were soaked under at least 7 cm of room-temperature water for up to 12 hours (as shown in Figure 2). This process ensured that each wood piece was thoroughly re-wetted prior to a given test.



Figure 2. Individual fuel pieces soaking under inches of water (left side) and view of wood pieces stacked before being held underwater (right side).

On the morning of the test day, wood was removed from the soaking bin and the outsides were dabbed dry with paper towels to ensure that a surface water coating was not present to interfere with the lighting of the pieces. The kiln-dried wood was also removed, and all pieces were re-weighed. These weights were used to calculate moisture contents of each of the individual pieces. The soaked fuel pieces were then further dried (if needed) in a kiln at just above 100 degrees Celsius and allowed to cool before testing to achieve target moisture contents.

The representative piece of kiln-dried wood was used with Equations 1-3 to estimate moisture content and target weight of each test piece, as shown below. The starting moisture content of the reference (kiln-dried) piece ($MC_{r,0}$) was determined following kiln drying according to equation 1, where the subscript r denotes 'reference', d denotes 'dried', and '0' represents the original value MC prior to kiln drying. In equation 1, m represents the mass of the fuel piece.

Kiln Dried Wood:
$$MC_{r,0} = \frac{m_{r,0} - m_{r,d}}{m_{r,d}}$$
 (1)

After the original moisture content of the test pieces were estimated using equation 1, the dry weights of the test pieces were estimated by applying equation 2, where the subscript t denotes 'test fuel', d denotes 'dried', r stands for 'reference', and '0' denotes the original value of the mass or moisture content.

Test Pieces:
$$m_{t,d} = \frac{m_{t,0}}{MC_{r,0}+1}$$
 (2)

Once the dry masses of the test fuel pieces were calculated using equation 2, equation 3 was used to determine how much water mass needed to be added to the test fuel pieces to achieve the target moisture contents ($MC_{t,w}$) of 5%, 15%, and 25%. In equation 3, the subscript t denotes 'test fuel', w denotes 'wetted', and d denotes 'dried'.

Test Pieces:
$$m_{t,w} = m_{t,d}(MC_{t,w} + 1)$$
 (3)

1.5 Sampling Setup and Emissions Measurement

A simplified graphic of the test setup is shown in Figure 3, including all instruments used for capturing emissions and their respective sampling locations relative to the main hood exhaust line.

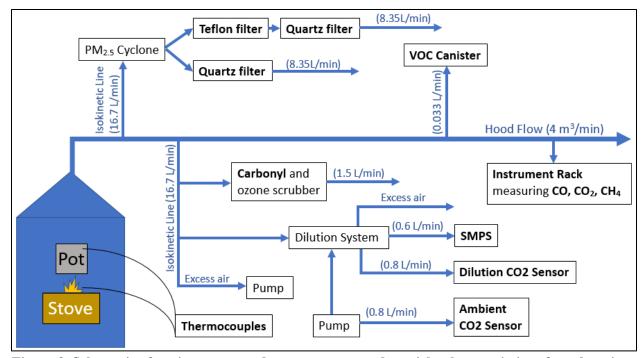


Figure 3. Schematic of testing setup used to capture gas and particle-phase emissions from burning wood in a rocket-elbow stove where bolded words represent sampling instruments

To collect particle emissions, two stainless-steel filter cartridge holders were placed downstream of a cyclone with a 2.5 μm cut point operating at 16.7 L/min of flow. Each filter line was operated at 8.35 L/min of flow. One stainless-steel filter cartridge holder contained a Teflon filter [PM2.5 Membrane, Polytetrafluoroethylene 46.2mm with support ring, 2.0um, #SF18040, Tisch] for gravimetric analysis, followed by a quartz filter [Tissuequartz, #2500QAT-UP, Pall Life Sciences] to correct for potential organic carbon (OC) adsorption artifacts. The other cartridge holder contained a single quartz filter for measurement of PM_{2.5} elemental carbon and organic carbon (EC/OC). Teflon filters were each pre-weighed on a microbalance [MX5, Mettler Toledo] three times and weights were averaged. Filters were kept in petri dishes until testing and were placed back into the filter weigh room immediately after testing to reach equilibrium with the room's temperature and relative humidity. Once equilibrium was achieved after at

least 12 hours, filters were reweighed. A lab blank was weighed for every set of filters to account for changes in scale-room conditions that might affect filter weights. After Teflon filters were post-weighed, filters were stored in a freezer at -80 °C.

Quartz filters were analyzed for elemental and organic carbon in an EC/OC analyzer [OC-EC Aerosol Analyzer, Sunset Laboratories Inc.] using the NIOSH 930 thermal protocol with a 270-helium step. 1.5 cm² punches were taken out of the filters and run through the analyzer. The front quartz filter that collected both gas- and particle-phase organic carbon was corrected using OC levels measured on the quartz-behind-Teflon filter, which only collected gas-phase organic carbon. After quartz filters were analyzed, they were stored along with the Teflon filters in the -80 °C freezer.

Gas-phase emissions included VOCs, (benzene, toluene, ethylbenzene, o-xylenes, and m+p-xylenes, grouped together as "BTEX"), select carbonyls (including acetaldehyde and formaldehyde,) CO, CO₂, and CH₄. To ensure that only emissions from the test fuel pieces were captured, valves to the filter cartridge holders and to the VOC canister [2L evacuated, electropolished canisters, University of California, Irvine] were not opened until the first set of test pieces were ignited in the stove. A placeholder dinitrophenylhydrazine (DNPH)-coated sampling cartridge [Sep-Pak DNPH-Silica Plus Short Cartridge, 350 mg Sorbent per Cartridge, 55-105 um Particle Size, Waters Corp.] for capturing carbonyls and ozone scrubber [Sep-Pak Ozone Scrubber Potassium Iodide, Plus Short Cartridge, 1.4 g, 55-105 um Particle Size, Waters Corp.] were also left in the line until sampling time, when the test DNPH cartridge was inserted. At the end of the test, the test DNPH cartridge was removed and the filter cartridge holder and VOC lines were closed before the fire burned out to limit non-representative emissions.

All other instruments were turned on, flow-checked, and sampling before the test started and continued after the test ended. Data for the continuous sampling instruments – namely the SMPS, CO analyzer [ULTRAMAT/OXYMAT 6, Siemens], and CO₂/CH₄ analyzer [ULTRAMAT 23, Siemens] were truncated to the correct times during data clean-up. Post-test flow checks were also conducted on the filter cartridge, isokinetic, and DNPH cartridge lines to ensure that lines were pulling emissions at expected rates

throughout the testing period. CO, CO₂, and CH₄ emissions were collected using Siemens ULTRAMAT analyzers that use nondispersive infrared (NDIR) sensor technology to quantify concentrations. Each gas analyzer was calibrated on the morning of each test day using those respective gases to span the instruments and using ultra-high purity N₂ as the zero gas. DNPH -derived carbonyls were detected by absorbance on an Agilent Model 1050 HPLC equipped with a diode array detector. BTEX VOCs were analyzed using gas chromatography (using flame ionization technology for benzene and toluene and mass spectrometry for ethylbenzene and xylenes).

1.6 Burning Procedure

For each test, the Envirofit G3300 stove was placed on a flat cement surface in the middle of the test hood. A pot containing 5 L of room-temperature water was placed on top of the stove for testing. Thermocouples connected to a real-time data logger [HH374 4-Channel Data Logger Thermocouple, OMEGA Engineering] were placed at the top of the stove and in the pot of water (approximately 6 cm below the surface of the water). A char collection pot and its lid were weighed for later use.

To light the stove, three shims [White Wood Shims, Nelson Wood Shims] were weighed and placed into the stove on top of a small wire grate to aid air circulation in the combustion chamber. Shims were then lit using a match and burned until approximately 3 cm of unburned wood was left on the shims. At this point, two pieces of test fuel were weighed for the last time immediately before feeding them into the stove combustion chamber with ~3 cm of overlap with the remaining shims. Once the first set (the two test pieces) were lit and burning without assistance from the shims, the lines to the filters, VOC canister, and DNPH cartridge were opened and this was considered the start of the emissions collection period. The two fuel pieces were then fed inward to sustain the flame in the combustion chamber and keep gas emissions as stable as possible. Once the first two test pieces were burned through with ~5 cm of unburned material remaining, the next set of two pieces were individually weighed and fed into the stove in the same manner and rate as the first set. This was repeated once more with a third set of two pieces. Once the third set of fuel was burned to the end of the pieces, the instrument lines were closed, the placeholder DNPH cartridge was reinserted, and this was considered the end of the emissions collection period.

At the point of the third set of pieces being burned to their ends, the pot with water was quickly removed and weighed to determine how much water had evaporated. That pot was then emptied into a sink and set aside. Immediately after weighing the water pot, unburned segments of fuel from the stove were placed into the previously-weighed char-pot with the lid closed to prevent further burning, and the pot with the unburned pieces was weighed. The char pot was then held inside the hood and the lid was lifted to allow smoke to escape. The remaining char and ash from inside the combustion chamber were then scooped into the char pot and it was re-weighed. The unburned pieces and char and ash were emptied into a designated bin and the stove was removed from the hood and placed in front of a fan to cool completely before the next test. Post-test flow checks were performed for the filter holders, DNPH cartridge, and isokinetic lines and were averaged with the pre-test flow checks to determine average flow rates to use in pollutant concentration calculations.

Between tests, filter holders, the VOC canister, and the carbonyl were changed out and all pre-test flow checks were performed. Enough time was allotted between tests for the stove to cool back down to ambient temperatures. Between tests, carbonyls and filter cartridges were placed into plastic bags to avoid picking up extra pollution from the room.

1.7 Data Analyses

The emissions collected during experimentation generated a large dataset. Excel 2016 (Microsoft Corp., Redmond, WA, USA) and Google Sheets (Google LLC., Mountain View, CA, USA) were used for tracking data input during testing, and R 3.3.1 (R Core Team, Vienna, Austria) was used for all data cleaning, processing, and analysis. Data cleaning involved cutting start-up and burn-out times out of the continuous sampling instruments, taking mean outputs of continuously-sampling instruments, calculating concentrations from filter areas and flow rates, and converting all emissions concentrations into mass-based emissions factors. To convert gases from parts per million by volume measurements to mass concentrations, equation (4) was used.

$$concentration \left[\frac{\mu g}{m^3}\right] = c = \frac{MW \left[\frac{g}{kmol_{gas}}\right] * ppmv \left[\frac{kmol_{gas}}{10^6 \, kmol_{air}}\right] * P \left[kPa\right]}{R_u \left[\frac{kPa*m^3}{kmol_{air}*K}\right] * T \left[K\right]} \tag{4}$$

Where "MW" is the molecular weight of the gas, "ppmv" stands for "parts per million by volume" and is the average number of moles of gas per megamole of air for each test, "P" is the room pressure, "R_u" is the universal gas constant, and T is the room temperature. To convert measured air concentrations to total grams of pollutant emitted, equation (4) was altered as shown in equation (5).

total mass emitted =
$$m_{tot_emitted} = c \left[\frac{\mu g}{m^3} \right] * t \left[min \right] * f_{hood} \left[\frac{m^3}{min} \right] * 10^{-6}$$
 (5)

Where "t" is the duration of sampling and "fhood" is the flow rate of the main exhaust line.

To convert the total mass of the pollutants to fuel mass-based emissions factors, equation (5) as altered as shown in equation (6).

$$EF\left[\frac{g_{pol}}{kg_{dryfuel}}\right] = \frac{total\ mass\ emitted}{m_{dry_estimate}} \tag{6}$$

Emissions factors based on energy delivered to the pot of water were also calculated, and those results are shown in the appendix.

Test-day background filter weights were used to correct for background concentrations of particles. The difference between filter weights measured before and after testing was converted into particle concentrations using equation (7).

PM2.5 concentration
$$\left[\frac{\mu g}{m^3}\right] = \frac{\Delta mass \left[\mu g\right]}{flow \left[\frac{m^3}{min}\right] * dur[min]}$$
 (7)

Particle size distributions from the SMPS were first normalized so they could be compared between tests at each moisture level and between moisture levels. Frequencies of each particle size bin were calculated by dividing the average number of particles in each bin across scans within a test by the total average number of particles produced during each test.

Measurements taken from the CO and CO₂ analyzers were used to calculate modified combustion efficiencies at each fuel moisture level using equation 8. Measured test concentrations of CO and CO₂ were corrected for average background levels in the lab over the eight test days to calculate MCE as shown in equation 8.

$$MCE = \frac{(CO_2)_{test} - (CO_2)_{bg}}{((CO_2)_{test} - (CO_2)_{bg}) + ((CO)_{test} - (CO)_{bg})}$$
(8)

Sample size was small, samples were independent of one another, and the data could be approximated as normal, so t-tests were used to compare emissions at the different moisture levels. Pairwise t-tests were run to examine differences in emissions for each fuel shape/moisture content level as a separate category and then with both shapes grouped together for each moisture content level. Paired instead of two-sample t-tests were used because differences between groups were of interest and the study was modeled like a within-subject design, where moisture levels were compared to one another.

CHAPTER 3. RESULTS

When evaluating the effects of moisture content on the measured emissions, split and milled woods were considered together, since pairwise t-tests run on each of the pollutants suggested that split vs. milled fuel created no significant difference of emissions at each moisture level (at the 95% confidence level).

1.8 Modified Combustion Efficiency

Trends of modified combustion efficiency (MCE) and fuel moisture content are shown in Figure 4. Measured MCE of the stove decreased significantly from the driest to the wettest fuel fuels (p < 0.01). No significant differences were seen between low and medium or between medium and high moisture fuels (p > 0.05), although a trend in reduced MCE with increasing MC is evident in Figure 4. Median MCEs at each moisture level were 98.4% (standard deviation (sd) = 0.60%), 97.5% (sd = 0.62%), and 97.2% (sd = 0.49%) at the low, medium, and high fuel moisture contents respectively.

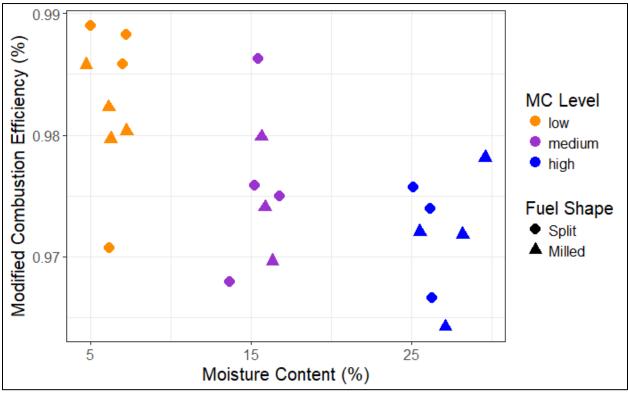


Figure 4. Modified combustion efficiency as it varies with moisture content of fuel.

1.9 Methane, Carbon Monoxide, and Carbon Dioxide

Trends in gas emissions with moisture content are shown in Figure 5. Methane, carbon monoxide, and carbon dioxide emissions factors all increased with increasing moisture content. Methane emissions factors doubled from low moisture content fuels (median = 1.82 g/kg, sd = 0.24 g/kg) to high moisture fuels (median = 3.83 g/kg, sd = 0.85 g/kg). Methane emissions were significantly different between all moisture levels (p = 0.017 low to medium, p = 0.02 medium to high, p < 0.01 low to high). The median CO emissions factors for the low, medium, and high fuel moisture levels were 22.2 g/kg (sd = 5.3 g/kg), 41.7 g/kg (sd = 8.0 g/kg), and 43.6 g/kg (sd = 8.8 g/kg), respectively, with the wettest fuels producing nearly twice the mass of CO per kg of fuel as the driest fuels. Carbon monoxide emissions were significantly different between low and medium levels (p < 0.01) and low to high levels (p < 0.01). Median CO₂ emissions ranged from 2221 g/kg (sd = 154 g/kg) for the low moisture content fuel to 2260 g/kg (sd = 121 g/kg) for the high moisture content fuel, which is less than a 2% difference. Carbon dioxide had no significant differences between any of the moisture levels.

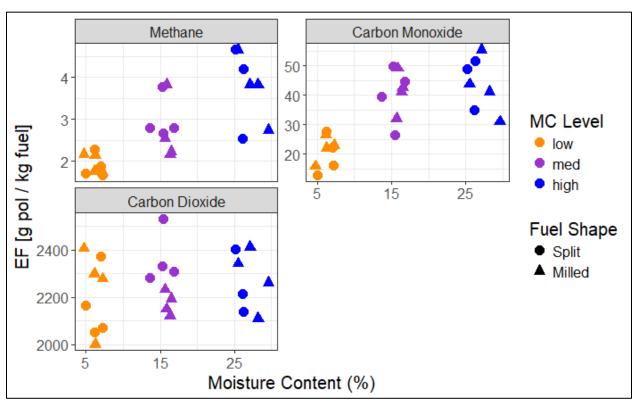


Figure 5. Emissions factors of methane, carbon monoxide, and carbon dioxide vs. fuel moisture content.

1.10 PM_{2.5}

Emissions factors of fine particulate matter increased significantly between all three moisture levels (p = 0.022 from low to medium, p < 0.01 from medium to high, and p << 0.01 from low to high fuels). Median emissions factors of $PM_{2.5}$ were 1.52 g/kg (sd = 0.38 g/kg) at the low moisture level, 2.35 g/kg (sd = 0.70 g/kg) at the medium level, and 4.36 g/ kg (sd = 1.2 g/kg) at the high level. High moisture-content fuel produced nearly 3 times as much particulate mass per kg of fuel than the low moisture content fuel. The trend of $PM_{2.5}$ emissions factors varying with fuel moisture is shown in Figure 6.

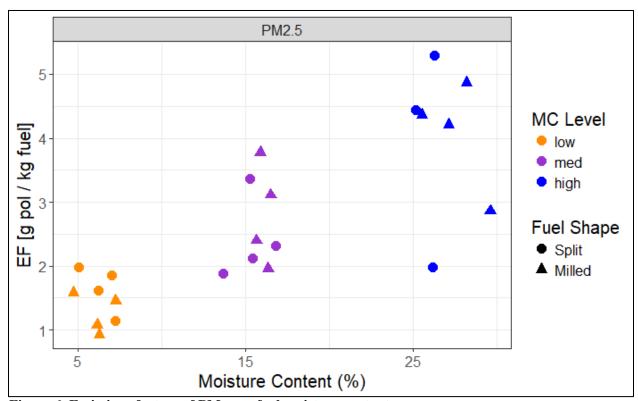


Figure 6. Emissions factors of PM_{2.5} vs. fuel moisture content.

1.11 Elemental and Organic Carbon

Median elemental carbon emissions factors decreased from 0.73 g/kg (sd = 0.2 g/kg) at the low moisture level to 0.33 g/kg (sd = 0.23 g/kg) at the medium level to 0.22 g/kg (sd = 0.05 g/kg) at the high level. The emissions factors were over 3 times greater at the low moisture level than at the high moisture level. Elemental carbon (EC) decreased significantly when moisture content was increased from low to medium levels (p < 0.01) and from low to high moisture levels (p << 0.01), with no significant difference between medium and high levels.

Median organic carbon emissions factors increased from 0.42 g/kg (sd = 0.10 g/kg) at the low moisture level to 1.09 g/kg (sd = 0.60 g/kg) at the medium level to 2.46 g/kg (sd = 0.59 g/kg) at the high level. Organic carbon emissions factors were nearly 6 times greater for the wettest fuels than for the driest fuels. Organic carbon (OC) significantly increased between all levels (p < 0.01 low to medium, p < 0.01 medium to high, and p << 0.01 low to high). EC and OC results are shown in Figure 7.

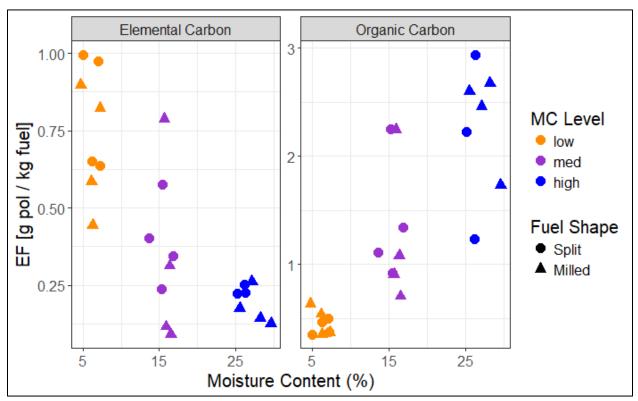


Figure 7. Emissions factors of EC and OC as they vary with fuel moisture.

Taking the ratio of elemental to organic carbon, significant decreases occurred between the low moisture fuel and medium moisture fuel (p < 0.01) and between low moisture fuel and high moisture fuel (p < 0.01). No significant difference was observed between medium and high moisture fuels. The EC/OC ratio decreased from a median of 1.12 to 0.07 when moisture content was increased from low to high levels. The trend of EC/OC ratio to moisture level is shown in Figure 8.

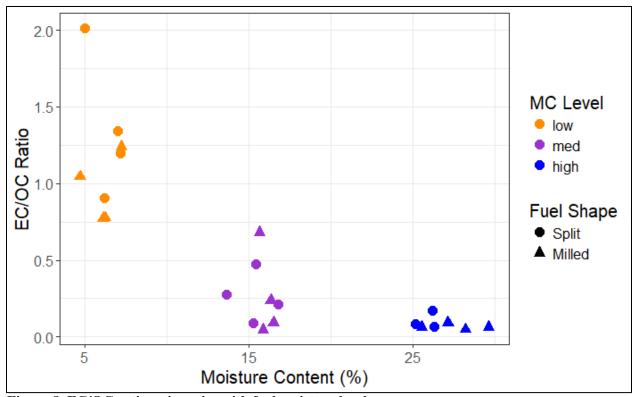


Figure 8. EC/OC ratio as it varies with fuel moisture level.

1.12 Carbonyls

A wide range of carbonyls were evaluated during this study, and their full results can be seen in Figure 14 in the Appendix. Acetaldehyde and formaldehyde results are reported in Figure 9 as acetaldehyde is possibly carcinogenic to humans and formaldehyde is a known carcinogen [19, 20]. Median acetaldehyde emissions factors increased 3.7 times from 5.3 mg/kg (sd = 1.41) mg/kg for low moisture fuel to 19.8 mg/kg (sd = 6.58 mg/kg) for high moisture fuels. Acetaldehyde emissions increased significantly from low to medium moisture levels (p < 0.01) and from low to high moisture levels (p << 0.01). Formaldehyde emissions increased significantly as moisture increased (p << 0.01 for low to medium, p < 0.01 for medium to high, and p << 0.01 for low to high). Formaldehyde emissions factors at the high moisture levels (median = 53.3 mg/kg, sd = 9.67 mg/kg) were over 3 times greater than emissions factors at the low moisture level (median = 17.2 mg/kg, sd = 4.18 mg/kg).

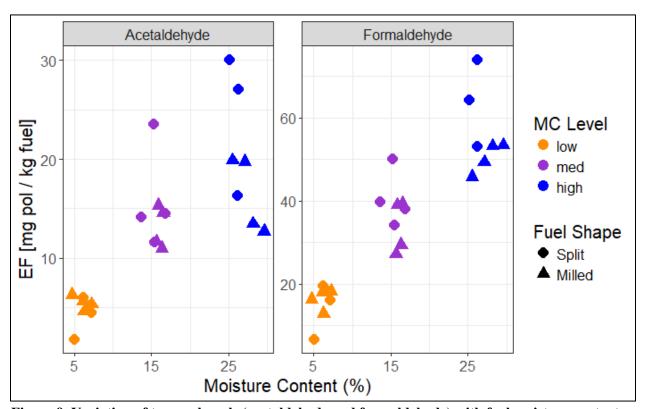


Figure 9. Variation of two carbonyls (acetaldehyde and formaldehyde) with fuel moisture content.

1.13 Volatile Organic Compounds

Benzene emissions factors increased significantly from low (median = 0.08 g/kg, sd = 0.067 g/kg) to high (median = 0.21 g/kg, sd = 0.062 g/kg) moisture levels (p = 0.018). Toluene emissions factors increased significantly between low (median = 0.02 g/kg, sd = 0.016 g/kg) and medium (median = 0.05 g/kg, sd = 0.025 g/kg) moisture levels (p = 0.020) and between low and high (median = 0.07 g/kg, sd = 0.018 g/kg) moisture levels (p < 0.01). Ethylbenzene emissions factors increased significantly between low (median = 0.003 g/kg, sd = 0.0019 g/kg) and medium (median = 0.007 g/kg, sd = 0.0026 g/kg) moisture levels (p = 0.037) and between low and high (median = 0.013 g/kg, sd = 0.0045 g/kg) levels (p < 0.01). M- and p-xylenes emissions factors increased significantly between low (median = 0.005 g/kg, sd = 0.003 g/kg) and medium (median = 0.011 g/kg, sd = 0.006 g/kg) moisture levels (p = 0.032) and between low and high (median = 0.023 g/kg, sd = 0.009 g/kg) levels (p < 0.01). O-xylenes emissions factors increase significantly between low (median = 0.002 g/kg, sd = 0.003 g/kg) and medium (median = 0.004 g/kg, sd = 0.003 g/kg) moisture levels (p = 0.018) and between low and high (median = 0.007 g/kg, sd = 0.003 g/kg) moisture levels (p = 0.018) and between low and high (median = 0.007 g/kg, sd = 0.003 g/kg) moisture levels (p = 0.018) and between low and high (median = 0.007 g/kg, sd = 0.003 g/kg) moisture levels (p < 0.018). Trends in BTEX emissions factors with fuel moisture content are shown in Figure 10.

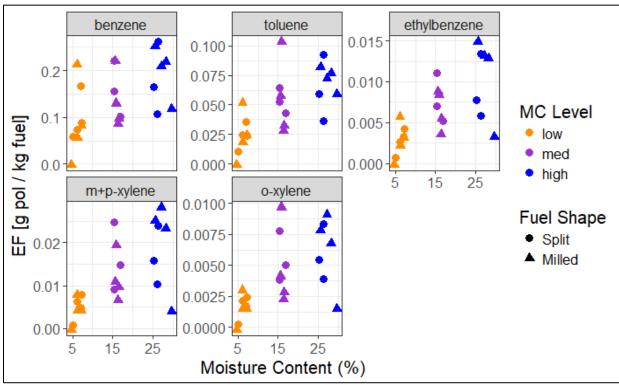


Figure 10. BTEX emissions factors as they vary with fuel moisture content.

1.14 Particle Size Distributions

Particle size distributions were similar between medium and high moisture level fuels, and slightly altered for dry fuels (Figure 11). When dry fuels were burned, a higher percentage of the total number of particles were larger than 150 nanometers in diameter than was the case for wetter fuels. All three fuel moisture levels produced ultrafine particles in greater numbers than larger particles. The size distributions of particles for the driest fuels were dominated by particles approximately 40 nm in diameter, while size distributions for medium and high moisture fuels were dominated by particles approximately 60-80 nm in diameter. Graphs depicting the variation of size distributions within each moisture content are shown in the appendix (Error! Reference source not found. through Error! Reference source not found.2).

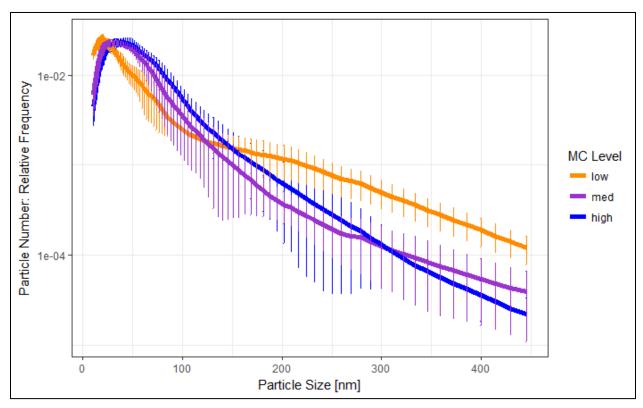


Figure 11. Average particle size distributions of three moisture levels for the range 10-490 nm, where error bars display ± 1 standard deviations from the mean.

1.15 Carbon Monoxide and PM_{2.5} Emissions Rates

Carbon monoxide was emitted at an average rate of 0.15 mg/min at low moisture level and 0.21 mg/min at the medium and high levels. The difference in rates means about 0.06 mg more CO were emitted every minute when wet fuel was burned compared to dry fuel. These differences are shown in Figure 12.

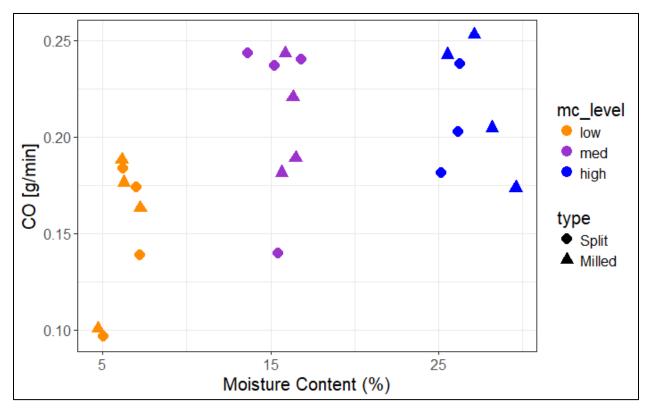


Figure 12. Rate of CO emissions at different fuel moisture levels.

 $PM_{2.5}$ was emitted at an average rate of 10.7 mg/min when dry fuel was burned, 13.5 mg/min when medium moisture fuel was burned, and 19.4 g/min when wet fuel was burned. The difference between burning dry and wet fuel was 8.7 g more $PM_{2.5}$ emitted every minute when wet fuel was burned. These differences are shown in Figure 13.

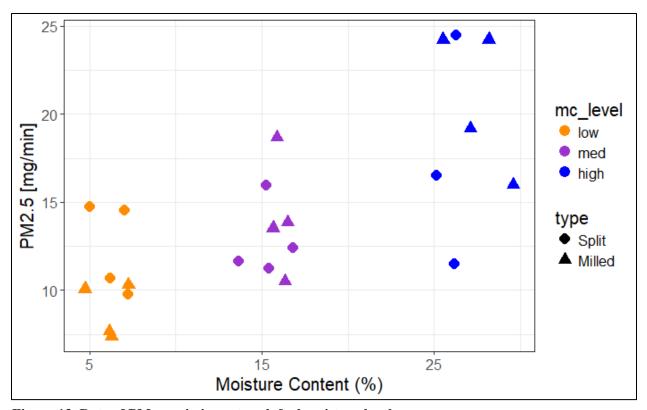


Figure 13. Rate of PM_{2.5} emissions at each fuel moisture level.

CHAPTER 4. DISCUSSION

All emissions factors, except those of EC and CO₂, increased with increasing moisture content between the 5% (low moisture) and 25% (high moisture) levels during this study. Modified combustion efficiency decreased with increasing moisture content of the Douglas fir in this study, contradicting findings by Jetter et al. that suggested a slight increase in MCE (from 96.7% to 97.7% when red oak was at 26% as opposed to 10% moisture content, although the trend seemed insignificant [2]. Many of the other stove/fuel combinations tested by Jetter et al. displayed either no significant differences between wet and dry fuel MCEs or MCEs that were slightly higher for dry fuel than for wet fuel [2]. These trends might be due to Jetter et al. using dry fuel as needed during the wet fuel tests to keep the flame burning, so average moisture contents of those tests were actually lower (around 20 - 25%) than the target 30% and therefore might not have been different enough from the "dry" fuel (target 10% MC) to display differences in MCE. By ensuring that all test pieces in the "wet" category of the tests performed in this study were above ~25% and that all pieces in the "dry" category for the tests were below ~8%, differences in MCE were more easily observed. Modified combustion efficiency is thought to decrease with an increase in moisture content since combustion temperatures are lowered when burning wetter fuel and more energy goes towards evaporating water out of the fuel than goes towards converting carbon in the wood to CO₂. The improved efficiency achieved when drying fuel to 5% as opposed to 25% MC suggests significant benefits to stove users.

Methane emissions increased with increasing moisture content, likely because CH₄ is produced mainly during pyrolysis of the wood, which takes longer for wet wood than for dry wood [21]. These findings contradicted findings by Jetter et al. and Bhattacharya et al., who found decreases in CH₄ and no change in CH₄ with increasing moisture content, respectively [2, 4]. Reasons for the discrepancies in CH₄ are unclear, but could be due to airflow into the stove, as this also affects pyrolysis rate. CO also increased with increasing moisture content in this study, which was opposite to differences observed by Jetter et al., which showed a decrease in CO from 35.7 g/kg to 26.4 g/kg when wood increased from 9 to 26% moisture content, although for multiple other stoves and fuels evaluated by Jetter et al. the trend in CO did increase [2].

Bhattacharya et al. found that CO emissions increased with increasing pine wood chip moisture content for the stoves tested (40.1 g/kg at 9.78% MC and 78.2 g/kg at 24.5% MC for the Indian "Harsha" stove, for example) [4]. Bhattacharya et al. concluded that the increase in CO emissions and slight decrease in CO₂ emissions at higher moisture contents were likely due to lower gas-phase oxidation rates caused by lower temperatures [4]. Physically, an increase in CO seems reasonable, as the decreased flaming ability of the fuel should make it difficult for all the carbon in the wood to be turned into CO₂, and should therefore go towards CO production and other carbon-containing emissions.

PM_{2.5} emissions factors agreed with findings by Shen et al., but trends were opposite to findings by Yuntenwi et al. and Jetter et al. who saw decreases in PM_{2.5} emissions factors even though they were also burning wood fuel in rocket-elbow stoves [2, 5]. The disagreements in trends might be due to the power at which stoves were operated, as high power produced larger emissions factors than low power. PM_{2.5} was mostly made up of organic carbon at the medium and high fuel moisture levels, and mostly made up of elemental carbon at the low moisture level. The remaining contributions to overall PM_{2.5} emissions likely came from ions, heavy metals, and inorganic material.

Since organic carbon is produced during smoldering at lower temperatures, while higher temperatures (produced at lower moisture contents) are needed for the formation of elemental carbon, trends in elemental and organic carbon seem reasonable. Organic carbon trends and levels agree with those observed by Shen et al. and Magnone et al, but elemental carbon trends were different from these studies [3, 9]. Since Shen et al. and Magnone et al. used different wood species (poplar and oak, respectively) and different wetting methods, the differences in EC trends might be explained by the temperatures and flame profiles achieved by the different wood species, as higher, flaming temperatures are needed for production of elemental carbon.

Emissions of two key carbonyls, acetaldehyde and formaldehyde, both increased with increasing fuel moisture content. The levels observed agreed with values observed by Zhang and Smith during comparison of carbonyls produced by various stove and fuel combinations [22]. The increase in both these carbonyls

and BTEX emissions with increasing moisture content of fuel could pose increased health risks to stove users who burn wood fuels in areas with relatively high EMC levels.

When emissions of CO and PM_{2.5} were evaluated on a rate basis, they could be compared to the Global Alliance for Clean Cookstoves' International Workshop Agreement (IWA) Tier system, which sets cutoff points for emissions to separate "cleaner" cookstoves from "dirtier" cookstoves [23]. When compared to the indoor air pollution tiers, emissions of PM_{2.5} per minute for dry and medium moisture fuel fell into the Tier 2 category (<17 mg/min) while emissions of PM_{2.5} for the wet fuel fell into the Tier 1 category (<40 mg/min). For CO emissions, all moisture levels fell into the Tier 4 (<0.42 g CO/min) category. For PM_{2.5}, the results of this study suggest that by drying fuel to approximately 5% moisture on a dry basis instead of 20%, the same stove operating under the same conditions can be classified as "cleaner" by international standards just by drying the fuel more completely. Thus, moisture content of fuel should be carefully controlled and monitored when certifying the emissions of a biomass cookstove.

With most emissions, except CO₂ and EC, increasing significantly from 5% to 25% moisture contents, estimates can be made for how regional variations in temperature and relative humidity might affect emissions from the example provided in the Introduction: if the same equilibrium moisture level Douglas fir was burned in a G3300 Rocket Elbow stove in Madras, India (10% EMC) and in Lagos, Nigeria (17.6% EMC), PM_{2.5} EFs could be estimated as increasing from ~2.25 g/kg to ~2.75 g/kg, CO EFs could increase from ~25 g/kg to ~40 g/kg, formaldehyde EFs could increase from ~25 mg/kg to ~45 mg/kg, and ethylbenzene could increase from ~4 mg/kg to ~6 mg/kg for example. In a "baseline" dry region such as Khartoum, Sudan, where the EMC is 4.8% in June, PM_{2.5} EFs would sit at ~1.25 g/kg, CO EFs would be at ~13 g/kg, formaldehyde EFs would be at ~14 mg/kg, ethylbenzene would be at ~ 2.5 mg/kg Similar variations in emissions would possibly also be observed at the same location if the stove was burned during different seasons with different relative humidity and temperature [14].

The overall physical effects of increasing fuel moisture content on stove emissions – reduced burning temperatures, longer burning times, more particulate matter production and smoldering – were all observed qualitatively while performing burn tests during this study, and agreed with results of similar studies [2,

10]. This suggests that the method of wetting wood under pressure in a tub of water overnight might be a viable option for future studies that aim to test fuel moisture in stoves or in general fire or combustion-related studies. Although *US EPA Method 28 – Certification and Auditing of Wood Heaters* does not allow dried wood to be re-wetted, the reasons behind this may not apply broadly, and may be limited to controlling stove certification tests by limiting the moisture range to between 19 and 26% on a dry basis [24]. Other sources explain that re-wetting mainly causes physical and mechanical properties to be altered, since below the fiber saturation point the moisture is absorbed by sponge-like cell walls that swell with increased moisture [18]. By re-wetting all the wood pieces from equilibrium moisture content, the chemical properties of all test pieces were as similar as possible (barring the innate differences in chemical properties of the wood throughout the log). Pilot testing suggested that moisture was evenly distributed radially and longitudinally throughout the wood after going through the soaking period.

Limitations of this work included only testing a single stove and wood species and utilizing an artificial re-wetting process. Testing more stoves and running a standard water boil test would help to apply the findings of this study to a broader range of situations. Performing many hypothesis tests during this study for each test to evaluate key results likely resulted in a few falsely low p-values due to the multiple testing issue. For this study, multiple testing is a minor issue, because consistent trends in the main effect estimates showed up across all the analyses for the various pollutants. However, there may a few "one-off" places where the t-tests produced inauthentically low p-values, and overall the chance that a null hypothesis (no differences between moisture levels) was incorrectly rejected may be higher than the 5% chance of falsely rejecting the null for any of the single tests. To build upon the findings of this study, test procedures could be repeated exactly as they were in this study, but instead of using artificial wetting, fuel from a single log could be cut and tested as it dried to the three moisture levels naturally. This could be done to assess how closely the re-wetting process from this study represents field wood at the same moisture levels achieved naturally, and could also allow for evaluation of wetter moisture levels.

CHAPTER 5. CONCLUSIONS

Drying Douglas fir wood from 25% to 5% significantly reduces many harmful emissions while increasing the modified combustion efficiency of a rocket elbow cookstove. In regions where the equilibrium moisture content is already higher than 10-15%, pre-drying the fuel before burning might be advantageous. Emissions reduced by drying include BTEX VOCs and carcinogenic carbonyls along with the more frequently-measured CO and PM_{2.5} emissions. Field studies comparing stove emissions in different regions of the world should account for moisture content of the fuel in that region, even if the same type of fuel is being used. Lab studies that aim to capture moisture content as a factor affecting emissions might benefit from the wetting process described in Methods, as it can save researchers a great deal of time. The decreased efficiency of the stove due to increased moisture is also relevant to those that use wood stoves for cooking or heating, as energy is lost to evaporating water out of the fuel during the burning process instead of going toward the desired purpose.

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APPENDICES

Four replicates of each moisture level/fuel shape combination were tested in a randomized matrix as shown below. The test codes below describe fuel shape (first letter: "milled" or "split"), moisture level (second letter: "low", "medium", or "high"), and replicate number. "BG1-8" represent the background tests for each test day.

Table 3. Randomized moisture content test matrix

Test Date	1st Test of Day	2nd Test of Day	3rd Test of Day	4th Test of Day
March 16, 2017	ML1	BG1	SL1	MH1
March 23, 2017	SM1	MM1	BG2	SH1
March 30, 2017	BG3	SM2	ML2	SL2
April 13, 2017	MM2	SH2	MH2	BG4
April 20, 2017	SL3	ML3	BG5	MM3
May 16, 2017	BG6	MH3	SH3	SM3
May 23, 2017	SH4	BG7	MM4	ML4
May 25, 2017	MH4	SL4	SM4	BG8

acetaldehyde acetone acrolein benzaldehyde 30 15 1.0 20 10 5 10 0.5 butyraldehyde butanone crotonaldehyde formaldehyde 3 6 60 2 2 mc_level 4 40 EF [g pol / kg fuel] 2 20 med high isovaleraldehyde hexaldehyde m_p_tolualdehyde m+p-xylene 0.5 type 0.4 0.02 20 field 2 0.3 0.01 0.2 lab 10 0.1 0.00 o_tolualdehyde valeraldehyde propionaldehyde methacrolein 1.2 40 1.5 10 0.9 30 0.6 1.0 20

Several additional carbonyls were evaluated during this study. They are shown in Figure 14.

Figure 14. Additional carbonyl emission factors vs. fuel moisture content.

med

high

Moisture Content (%)

10

0

low

high

med

0.5

Energy-based emissions factors were calculated for PM_{2.5}, CH4, CO, and CO₂, acetaldehyde, formaldehyde, benzene, ethylbenzene, toluene, elemental carbon, and organic carbon. The emissions factors are graphed in Figure 15 through Figure 19. Normalized particle size distributions were calculated for each moisture level and the results are shown in Figure 20 through Figure 22.

0.3

0.0

med

high

med

high

Energy-based emissions factors were calculated by dividing the mass of each pollutant emitted per test by Equation (9), which describes the MJ of energy delivered to the pot on the stove.

$$MJ_{delivered} = MJ_{heat} + MJ_{evap} = (m_{h2o} * c_{h2o} * \Delta T) + (m_{evap} * h_{fg})$$
 (9)

Where m_{h2o} is the initial mass of water in the pot, c_{h2o} is the specific heat, ΔT is the increase in the temperature, m_{evap} is the mass that evaporated, and h_{fg} is the heat of vaporization of the water.

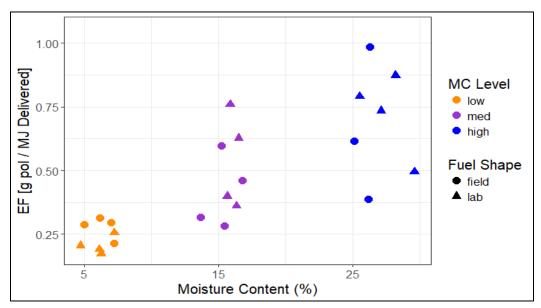


Figure 15. Energy-based emissions factors of $PM_{2.5}$ at three moisture levels.

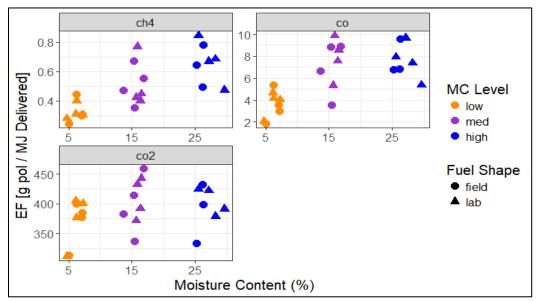


Figure 16. Energy-based emissions factors of CH₄. CO, and CO₂ at three moisture levels.

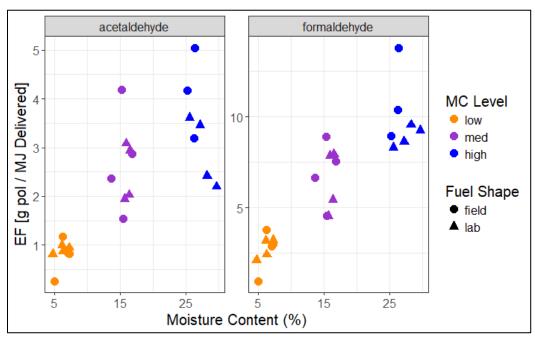


Figure 17. Energy-based emissions factors of acetaldehyde and formaldehyde at three moisture levels.

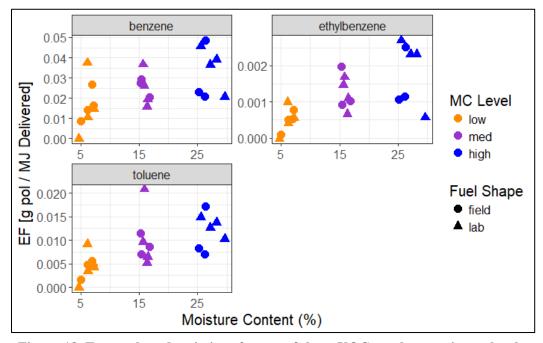


Figure 18. Energy-based emissions factors of three VOCs at three moisture levels.

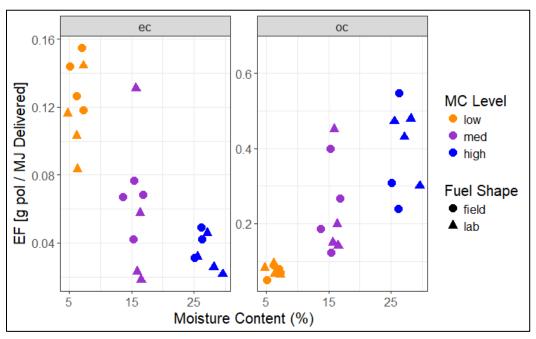


Figure 19. Energy-based emissions factors for elemental and organic carbon at three moisture levels.

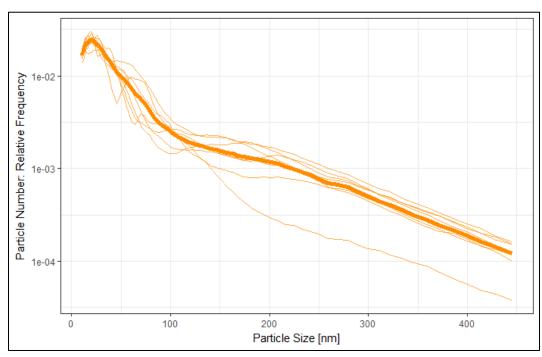


Figure 20. Relative size distribution of particles for dry moisture fuels with the average across tests in bold.

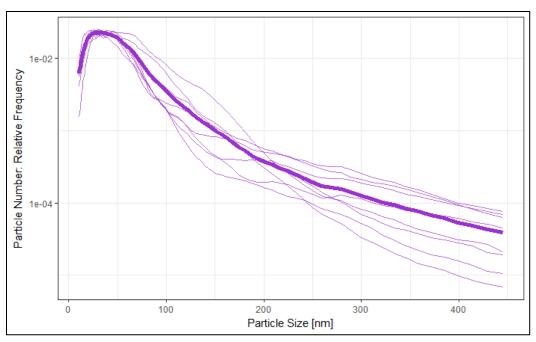


Figure 21. Relative size distribution of particles for medium moisture fuels with the average across tests in **bold**.

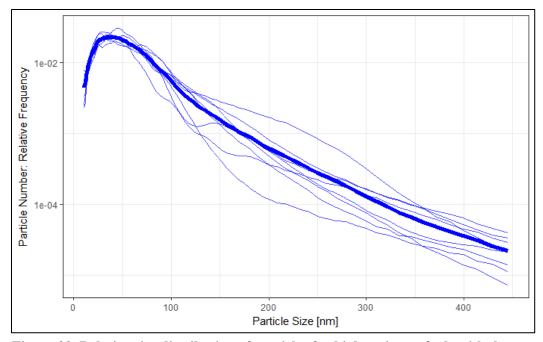


Figure 22. Relative size distribution of particles for high moisture fuels with the average across tests in **bold**.